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Article

The Effects of pH Change through Liming on Soil N₂O Emissions

Muhammad Shaaban ^{1,2}, Yupeng Wu ¹, Lei Wu ³, Ronggui Hu ^{1,*}, Aneela Younas ⁴, Avelino Nunez-Delgado ^{5,*}, Peng Xu ¹, Zheng Sun ^{6,7}, Shan Lin ¹, Xiangyu Xu ¹ and Yanbin Jiang ¹

¹ College of Resources and Environment, Huazhong Agricultural University, Wuhan 430070, China; shabanbzu@hotmail.com (M.S.); wyp19851205@mail.hzau.edu.cn (Y.W.); xupengstu192@163.com (P.X.); linshan@mail.hzau.edu.cn (S.L.); xuxiangyu2004@sina.com (X.X.); jiangyanbin@mail.hzau.edu.cn (Y.J.)

² Department of Soil Science, Faculty of Agricultural Sciences and Technology, Bahauddin Zakariya University, Multan 60800, Punjab, Pakistan

³ Institute of Agricultural Resources and Regional Planning, Chinese Academy of Agricultural Sciences, Beijing 100081, China; wuleies@163.com

⁴ College of Plant Science and Technology, Huazhong Agricultural University, Wuhan 430070, China; aneelayounas1@yahoo.com

⁵ Department of Soil Science and Agricultural Chemistry, Engineering Polytechnic School, Campus University, University Santiago de Compostela, 27002 Lugo, Spain

⁶ CNRS, EPHE, Sorbonne Universités, UPMC University Paris 06, UMR 7619 METIS, 4 Place Jussieu, CEDEX 05, 75252 Paris, France; zheng.sun@sorbonne-universite.fr

⁷ Geosciences Division, IFP Energies Nouvelles, 1 et 4 Avenue de Bois-Préau, CEDEX 92852 Reuil-Malmaison, France

* Correspondence: rg.hu@mail.hzau.edu.cn (R.H.); avelino.nunez@usc.es (A.N.-D.)

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Abstract: Nitrous oxide (N₂O) is an overwhelming greenhouse gas and agricultural soils, particularly acidic soils, are the main source of its release to the atmosphere. To ameliorate acidic soil condition, liming materials are added as an amendment. However, the impact of liming materials has not been well addressed in terms of exploring the effect of soil pH change on N₂O emissions. In the present study, a soil with pH 5.35 was amended with liming materials (CaMg(CO₃)₂, CaCO₃, Ca(OH)₂ and CaO) to investigate their effects on N₂O emissions. The results indicate that application of liming materials reduced the magnitudes of N₂O emissions. The maximum reduction of soil N₂O emissions took place for Ca(OH)₂ treatment when compared to the other liming materials, and was related to increasing soil pH. Mineral N, dissolved organic C, and microbial biomass C were also influenced by liming materials, but the trend was inconsistent to the soil pH change. The results suggest that N₂O emission mitigation is more dependent on soil pH than C and N dynamics when comparing the different liming materials. Moreover, ameliorating soil acidity is a promising option to mitigate N₂O emissions from acidic soils.

Keywords: lime; mineral nitrogen; soil pH; organic carbon; microbial biomass; N₂O

1. Introduction

Soil acidity is a master variable that hinders plant growth by limiting nutrient availability and thus impacts both the quantity and quality of crops. Soil acidification occurs very slowly naturally as soil is weathered, but this process is accelerated by intensive agriculture [1,2]. Soil acidity is expressed in terms of pH, and its extent and degree impact a wide range of soil biogeochemical properties. Soil acidity also has marked effects on soil microbial communities and their pertinent processes.

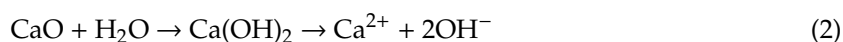
Soil acidification is a natural and very slow process that takes over hundreds of years to develop. However, it may reach its greatest expression within a few years under intensive agricultural practices and in humid regions where rainfall is sufficient to leach down the nutrients [3]. Thus, although most processes developing soil acidification are natural, anthropogenic activities have a major impact on some of them. In fact, several reasons may contribute to soil acidification and excessive use of nitrogen (N) is one of them [3].

To obtain high crop production in intensive crop-growing areas, excessive application of N fertilizers has been carried out for years, but when it is excessive, it leads to soil acidification [4]. According to estimations [3], the application of nitrogen fertilizer in arable lands of China usually ranges from 200 to 500 kg N ha⁻¹ yr⁻¹. Aside from the beneficial effects of high N fertilizer application, devastating impacts and environmental risks have also been observed including eutrophication, nitrous oxide (N₂O) emissions, and soil acidity [5,6]. Researchers have demonstrated that nitrate and ammonium applied to soils can generate 20 to 33 kmol hydrogen ions (H⁺) ha⁻¹ yr⁻¹ under exhaustive growing systems [3]. This indicates that the application of N can drive soil acidification.

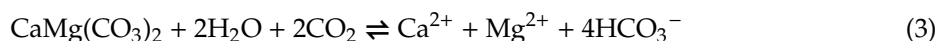
Soil acidity can be offset with alkaline materials that provide conjugate bases such as CO₃²⁻ and OH⁻ of weak acids. These anionic bases react with H⁺ and form weak acids. For example:



Generally, liming materials are applied in the forms of hydroxides or oxides containing magnesium (Mg) or calcium (Ca), which form hydroxide ions in water.



Most liming materials, whether they are carbonates, hydroxides, or oxides, react with CO₂ and H₂O to generate bicarbonates (HCO₃⁻) when added to acidic soils. As a result, partial pressure of CO₂ in the soil is high enough to proceed such reactions forward, for instance:



The resultant bicarbonates, Ca and Mg, counteract the acidity.

Liming acidic soils not only raises soil pH, but also alters biochemical processes and nutrient cycling. The rise in soil pH following lime application substantially triggers the N transformation processes [7], markedly controls the microbial processes of nitrification and denitrification, and thus influences N₂O production and emission. However, the subsequent effects of lime application on N₂O emissions is ambiguous and contrary hypotheses have been proposed by the scientific community. For instance, a laboratory incubation study proposed that increasing soil pH may substantially decrease emissions of N₂O from acidic agricultural soils [8]. In contrast, some scientists have reported that lime application and subsequent rise in soil pH caused increased soil N₂O emissions from arable acidic soils [9,10].

Keeping in mind the importance of liming acidic soils, we hypothesized that the application of lime materials can trigger N transformations following soil pH change and subsequently influence N₂O emissions in a way that would be interesting to further elucidate. Thus, the current study was conducted with the aim to examine and shed further light on the pH change effects of various liming materials on N₂O emissions from acidic agricultural soils.

2. Materials and Methods

2.1. Soil and Liming Materials

Soil was obtained from a rapeseed-rice cropping system, located in Xianing (a city of central China; 29°88'209'' N, 114°39'416'' E). According to Soil Survey staff [11], the soil is classified as Ultisol.

Soil (0–20 cm) was sampled from the selected field after rice harvest from multiple-points. A composite soil sample was made by mixing subsamples. Plant residues (straw and roots) were separated from soil. After shifting in the laboratory, soil was dried in the open air, crumbled, and then sieved through a 2 mm sieve. The basic soil chemical and physical analysis [12] was performed prior to onset of the experiment. Soil texture was silty clay loam. The main characteristics of soil are given in Table 1. Different liming materials (dolomite $\text{CaMg}(\text{CO}_3)_2$, calcium hydroxide $\text{Ca}(\text{OH})_2$, calcium carbonate CaCO_3 , and calcium oxide CaO) used in the present study were purchased from Xinjing Chemicals Co. Ltd. (Xiaogan, Hubei, China).

Table 1. Some selected physical and chemical characteristics of the tested soil.

$\text{pH}_{(\text{H}_2\text{O})}$	Total C (g/kg)	Total N (g/kg)	$\text{NO}_3^- \text{N}$ (mg/kg)	$\text{NH}_4^+ \text{N}$ (mg/kg)	Bulk Density (g/cm^3)	Cation Exchange Capacity ($\text{cmol}_\text{c}/\text{kg}$)
5.35	11.5	1.2	15.6	46.9	1.39	10.8

2.2. Experimental Setup for Collection and Analysis of N_2O

Initially, air dried soil without any amendment was incubated in a plastic (polyethylene terephthalate) tub with 20% gravimetric water content (60% water filled pore space) at a temperature of $25 \pm 1^\circ\text{C}$ for one week (Figure 1). After one week of initial incubation, incubated wet soil (100 g on dry basis) from the tub was placed in glass jars. Liming materials were added separately to the soil. The application dose of each liming material was 1 g kg^{-1} dry soil with a particle size $\leq 0.3 \text{ mm}$. Treatments for the present study were as follows: (i) $\text{Ca}(\text{OH})_2$, (ii) CaO , (iii) CaCO_3 , (iv) $\text{CaMg}(\text{CO}_3)_2$, and (v) control (soil without any amendment). Each treatment had three replicates. Treated soils in jars were placed in an electric-automated chamber (S-400-HP) and incubated at $25 \pm 0.5^\circ\text{C}$ in the dark for four weeks (28 days). During incubation, a plastic sheet with pin holes (about 30) was used on the top of each jar to reduce water loss, but permit gas exchange. Soil water content in each jar was sustained at 20% throughout the study by weighing jars and refilling with distilled water on a daily basis.

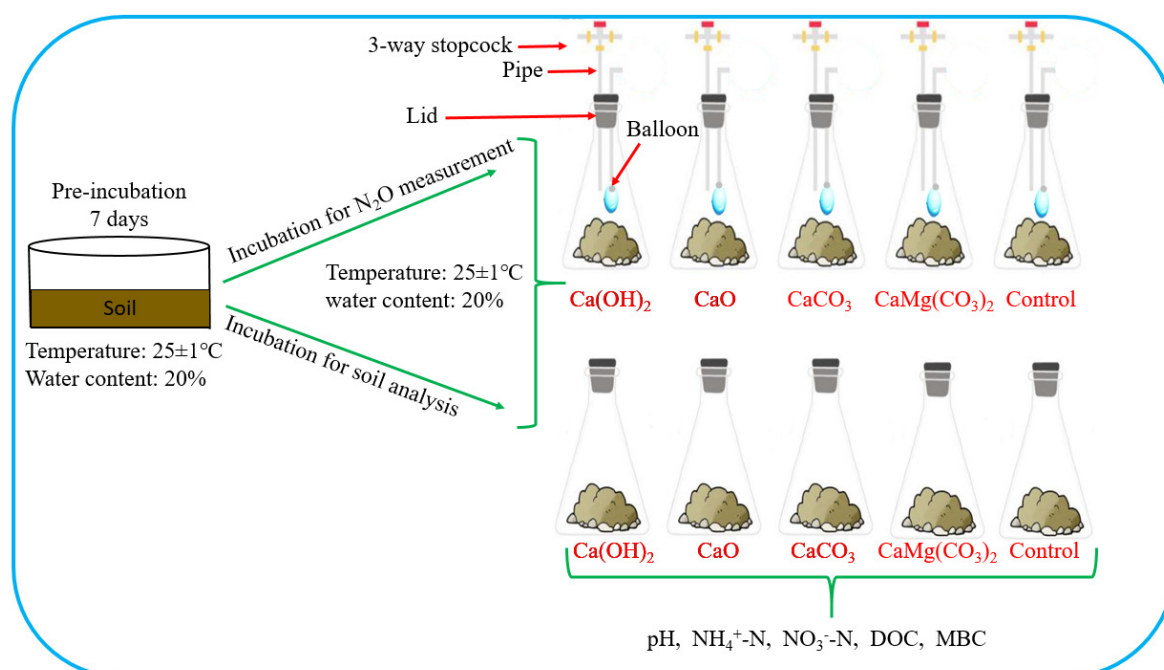


Figure 1. Schematic diagram of the experimental setup.

Gas from the headspaces of jars, equipped with air-tight lids holding a 3-mm diameter pipe, was collected at days 1, 3, 5, 7, 10, 13, 16, 20, 24, and 28 using a special air-tight syringe made for sampling purposes. On gas sampling day, the tops of jars were uncovered prior to gas sampling and soil in the jars was allowed to be exposed to ambient air for 30 to 40 min. After that, the jars were closed with air-tight lids and gas samples were taken immediately after closure to know the initial concentration of gas in the jars. Another gas sample from headspace was collected after 60 min to know the change in gas production. The gas samples were analyzed for N₂O concentration using a gas chromatograph system (7890A, Agilent technology, Santa Clara, CA, USA). The concentration of N₂O in the gas sample was calculated using the equation as given below [13].

$$F = \rho \times V/W \times \Delta c/\Delta t \times 273/(T + 273) \quad (4)$$

In Equation (1), F denotes the rate of N₂O–N emission ($\mu\text{g kg}^{-1} \text{h}^{-1}$); ρ denotes the density (kg m^{-3}) of N₂O gas; V denotes the volume (m^3) of headspace of jars; W denotes soil weight (kg); Δc denotes change in gas concentration during closure time of jars; Δt denotes the time period of closure (h) of the treatment jars; and T denotes the temperature at which the experiment was conducted (25°C).

The cumulative emissions of N₂O ($\mu\text{g kg}^{-1}$) for the whole period of study were calculated based on the following formula [14].

$$\text{Cumulative N}_2\text{O emission} = \sum_{i=1}^n (R_i \times 24 \times D_i) \quad (5)$$

where R_i is the N₂O emission rate ($\mu\text{g kg}^{-1} \text{h}^{-1}$); D_i are days between the sampling periods; and n is the number of samples.

2.3. Experimental Setup for Soil Analysis

A separate experiment to that for gas analysis was concurrently performed for soil analysis. Treatments, pre-incubation period, temperature, and moisture conditions for the soil analysis study were identical as that for the gas analysis setup. After pre-incubation, a weight of 200 g soil was incubated after being placed in 1000 mL beakers. Soil sub-samples from jars were taken after one day of imposing treatments and then on a weekly basis over 28 days.

For pH determination of the soil-sub samples, a soil slurry was made by performing a 1:2.5 ratio suspension of soil:distilled water [12]. The slurry was shaken in an orbital shaker for 40 min, and the pH was tested using a pH-meter (2FPHS, Wincoms Co. Ltd., Shanghai, China) after 30 min of shaking. Soil was subjected to specific extraction for the subsequent determination of the mineral contents of soil nitrogen (NO_3^- -N and NH_4^+ -N) by adding 1 M KCl (5 mL for 1 g soil), shaking for 60 min, and subsequently using a flow injector system analyzer (SEAL Co. Ltd., Henstedt-Ulzburg, Germany) [15]. Chloroform fumigation specific extraction method was adopted for testing microbial biomass C [16]. Dissolved organic C content in soil was determined by extracting the soil with distilled water (1:5, soil:distilled water) and using Elementar system analysis (Vario, Elementar-CN, Hanau, Germany).

2.4. Data Analysis

Data pertinent to soil and gas parameters were analyzed using Analysis of Variance (ANOVA) one-way analysis of variance. Tukey's test was employed to identify significant differences for treatments of their mean results. The Kolmogorov–Smirnov test for the normality distribution of variables was performed before proceeding further for ANOVA [17]. All data were statistically evaluated using Windows-based software Statistical Package for the Social Sciences (SPSS) Statistics 23.

3. Results

3.1. Soil pH

Soil pH was statistically significantly ($p \leq 0.01$) different among the treatments of liming materials. Soil pH before the immediate day of adding liming materials was 5.35, and liming of soil rapidly increased pH (Figure 2). On day 1, soil pH in all treatments was substantially higher than that of the control and thereafter continued to gradually increase up until the end of the study. The highest value of soil pH corresponded to $\text{Ca}(\text{OH})_2$ treatment on day 28. The pH values were 7.21, 6.99, 6.70, 6.43, and 5.30 for $\text{Ca}(\text{OH})_2$, CaO , CaCO_3 , $\text{CaMg}(\text{CO}_3)_2$, and the control, respectively, on day 28 of the experiment.

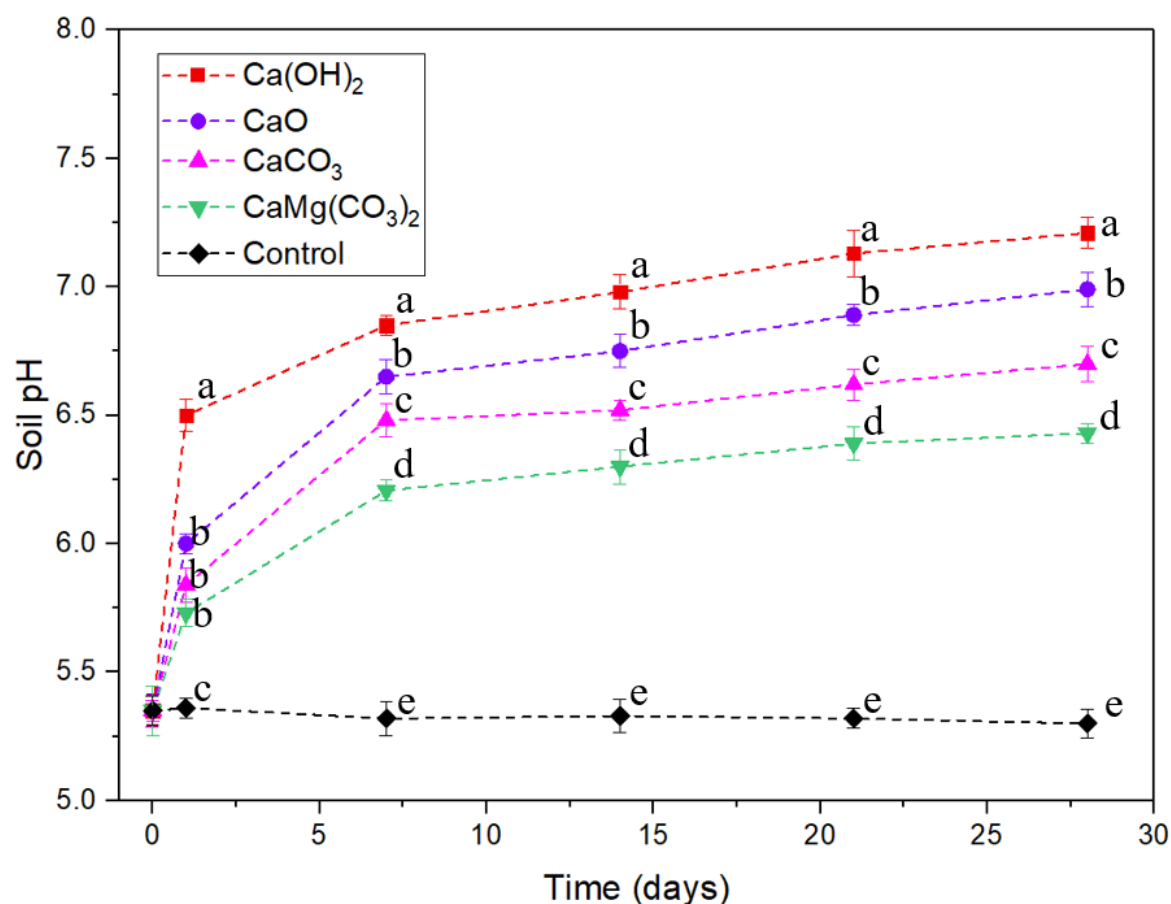


Figure 2. Dynamics of soil pH following the application of liming materials. Vertical bars denote the error bars of the mean of three replicates. Values at the same time followed by different letters indicate significant differences between different treatments ($p < 0.05$).

3.2. Soil Mineral-N ($\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$)

Soil $\text{NH}_4^+\text{-N}$ concentrations were highly and significantly ($p \leq 0.01$) influenced by the addition of liming materials. $\text{NH}_4^+\text{-N}$ concentration before the immediate day of adding liming materials was 35 mg kg^{-1} , whereas the addition of liming materials caused diverse patterns of $\text{NH}_4^+\text{-N}$ concentrations (Figure 3). The CaCO_3 , $\text{CaMg}(\text{CO}_3)_2$ and control treatments showed continuous decline of $\text{NH}_4^+\text{-N}$ concentrations throughout the study period. However, $\text{NH}_4^+\text{-N}$ concentration in the $\text{Ca}(\text{OH})_2$ and CaO treatments declined on day 1 of the onset of the study, increased on day 2, and afterward speedily decreased throughout until the end of the experiment. The $\text{NH}_4^+\text{-N}$ concentrations were 9.1, 10.9, 6.8, 12.0, and 20.2 mg kg^{-1} in the $\text{Ca}(\text{OH})_2$, CaO , CaCO_3 , $\text{CaMg}(\text{CO}_3)_2$ and control treatments, respectively, on day 28 of the study.

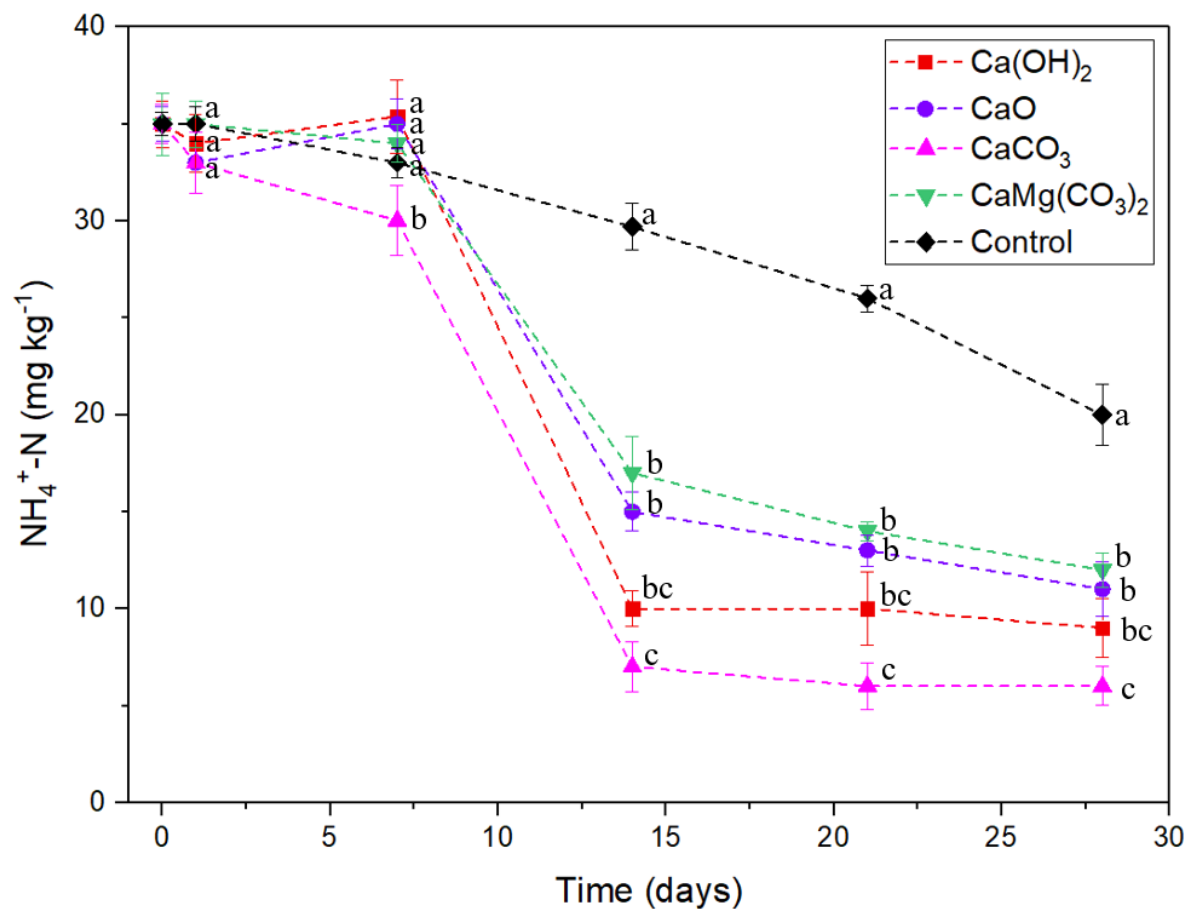


Figure 3. Variations in soil ammonium concentrations following the application of liming materials. Vertical bars denote the error bars of the mean of three replicates. Values at the same time followed by the different letters indicate significant differences between different treatments ($p < 0.05$).

The amendment of liming materials significantly ($p \leq 0.01$) augmented soil NO_3^- -N concentrations (Figure 4). The trend of increase of NO_3^- -N concentration kept continuing in all treatments until the end of the study. The maximum concentration of NO_3^- -N was observed in the CaCO_3 treatment on day 28 of the study. The NO_3^- -N concentrations were 60.2, 52.4, 75.1, 50.0, and 43.9 mg kg^{-1} in the Ca(OH)_2 , CaO , CaCO_3 , $\text{CaMg(CO}_3)_2$, and control, respectively, on day 28 of the study.

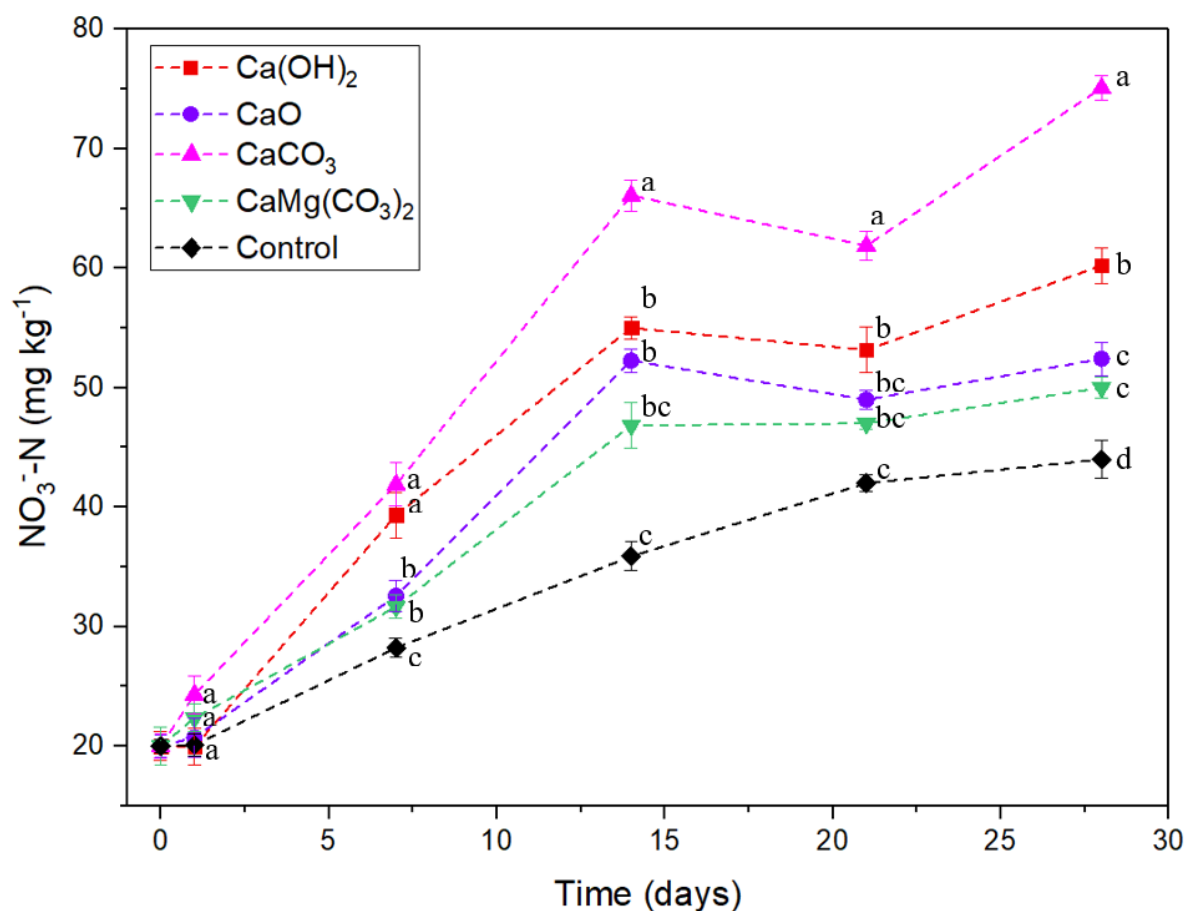


Figure 4. Variations in soil nitrate concentrations following the application of liming materials. Vertical bars denote the error bars of the means of three replicates. Values at the same time followed by different letters indicate significant differences between different treatments ($p < 0.05$).

3.3. Dissolved Organic C and Microbial Biomass C

Addition of liming materials significantly ($p \leq 0.01$) impacted the microbial biomass C (MBC) as well as dissolved organic C (DOC) in soil. Before the addition of liming materials, the DOC content was 25 mg kg^{-1} and instantly increased on day 1 in all treatments, except for the control (Figure 5). The DOC contents reached maximum values of 38.5, 33.2, 30.1, 28, 24.9 mg kg^{-1} on day 7 in the Ca(OH)_2 , CaO, CaCO_3 , $\text{CaMg(CO}_3)_2$, and control treatments, respectively, and afterward declined until the end of the study.

In the case of MBC contents, all the liming treatments showed an increment on day 1, while a divergent trend was observed afterward (Figure 6). Only Ca(OH)_2 treatment showed a rise in MBC content after day 1, reached the maximum at 59 mg kg^{-1} on day 14, and after that gradually declined and reached 49.1 mg kg^{-1} at the end of the experiment, whereas MBC contents decreased in all other treatments of CaO, CaCO_3 , and $\text{CaMg(CO}_3)_2$ as well as the control over the entire study period.

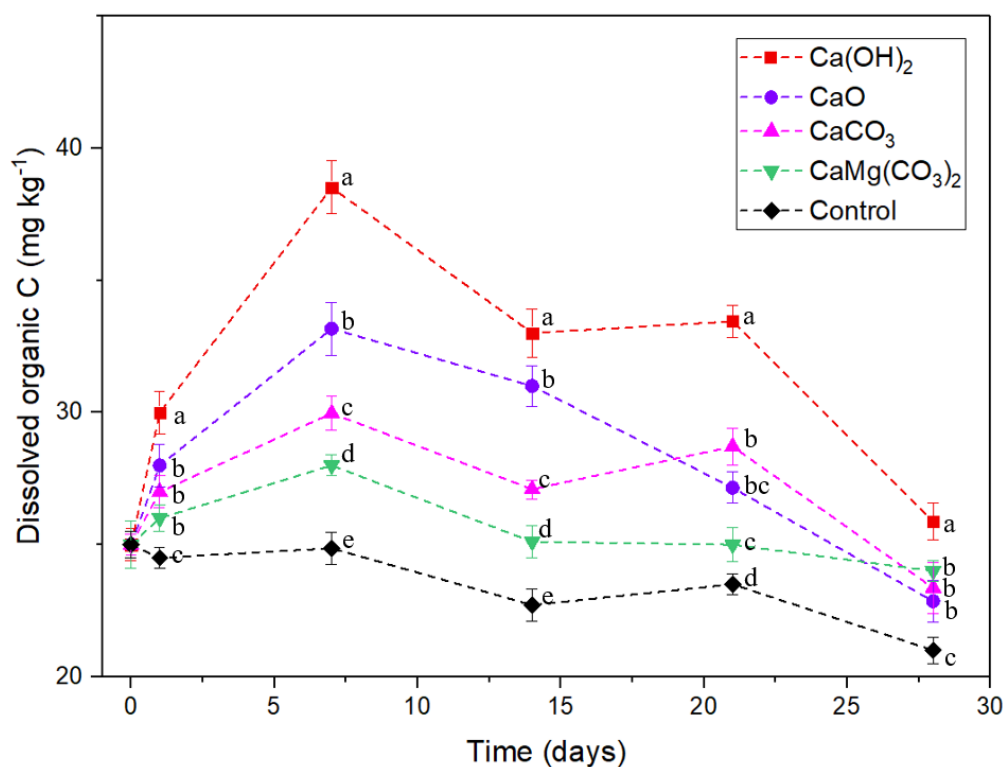


Figure 5. Fluctuations in soil dissolved organic carbon following the application of liming materials. Vertical bars denote error bars of mean of three replicates. Values at the same time followed by different letters indicate significant differences between different treatments ($p < 0.05$).

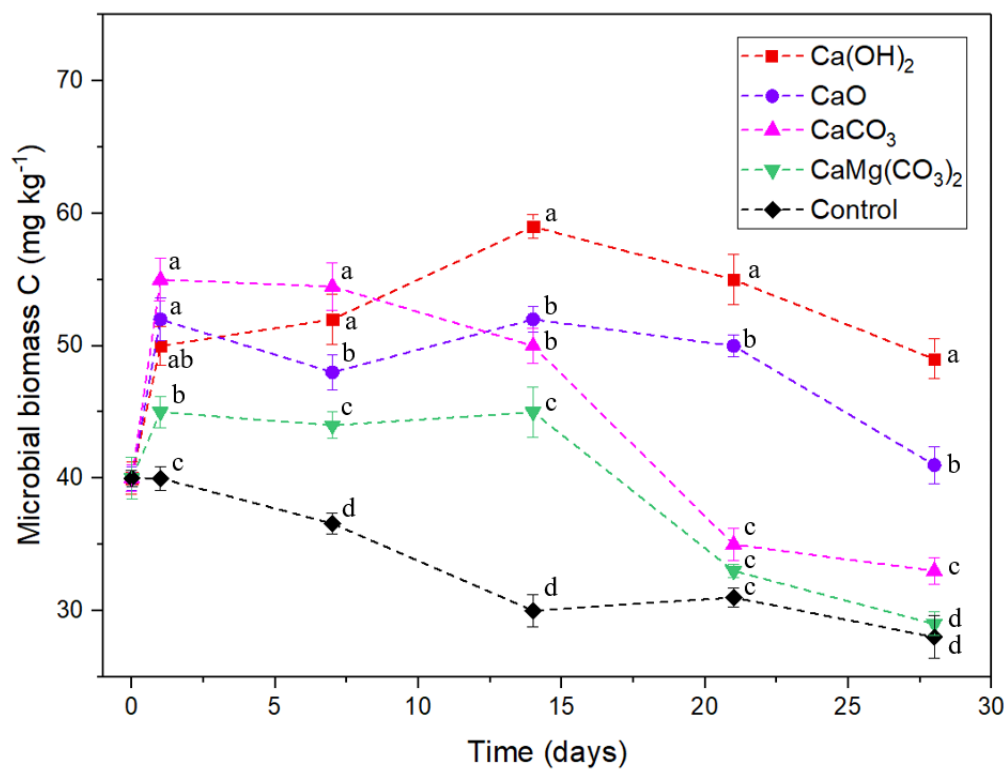


Figure 6. Fluctuations in soil microbial biomass carbon following the application of liming materials. Vertical bars denote the error bars of the means of three replicates. Values at the same time followed by different letters indicate significant differences between different treatments ($p < 0.05$).

3.4. Nitrous Oxide (N₂O) Emissions

Nitrous oxide emissions were significantly ($p \leq 0.01$) affected by the application of liming materials. The N₂O emission rate and the cumulative soil N₂O emissions ($329.52 \mu\text{g kg}^{-1}$) were highest in the control among all the treatments (Figures 7 and 8). The N₂O emissions increased on day 1 following the addition of liming materials, and then started to decline, with variant magnitudes (Figure 7). The decrease in N₂O emissions was sharper in Ca(OH)₂ than that of the other liming treatments, and indeed in the control. The lowest emission rate and cumulative N₂O emissions were observed in the Ca(OH)₂ treatment. The cumulative N₂O emissions in the Ca(OH)₂, CaO, CaCO₃, CaMg(CO₃)₂, and control treatments were 208.08, 237.60, 261.72, 287.64, and $329.52 \mu\text{g kg}^{-1}$, respectively (Figure 8).

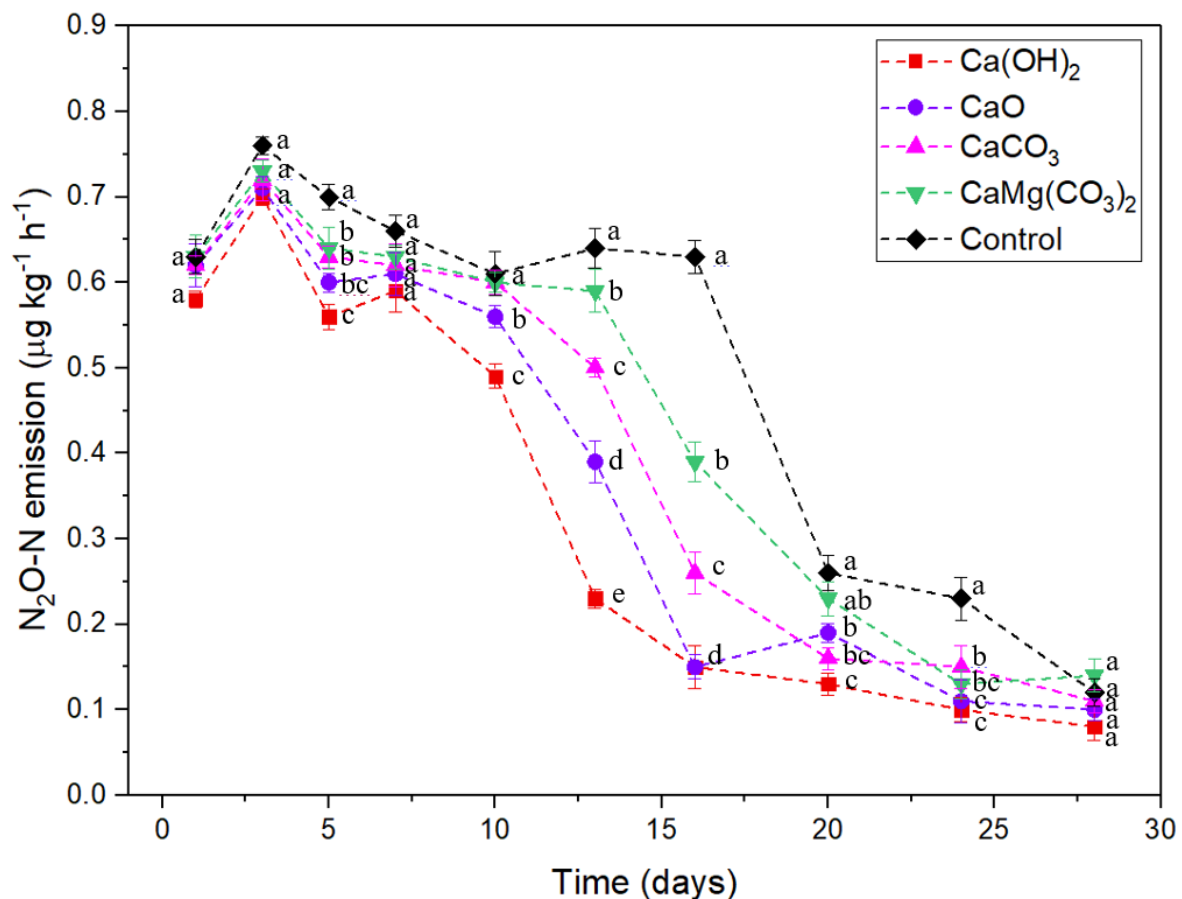


Figure 7. Soil N₂O emissions following the application of liming materials. Vertical bars denote the error bars of the means of three replicates. Values at the same time followed by different letters indicate significant differences between different treatments ($p < 0.05$).

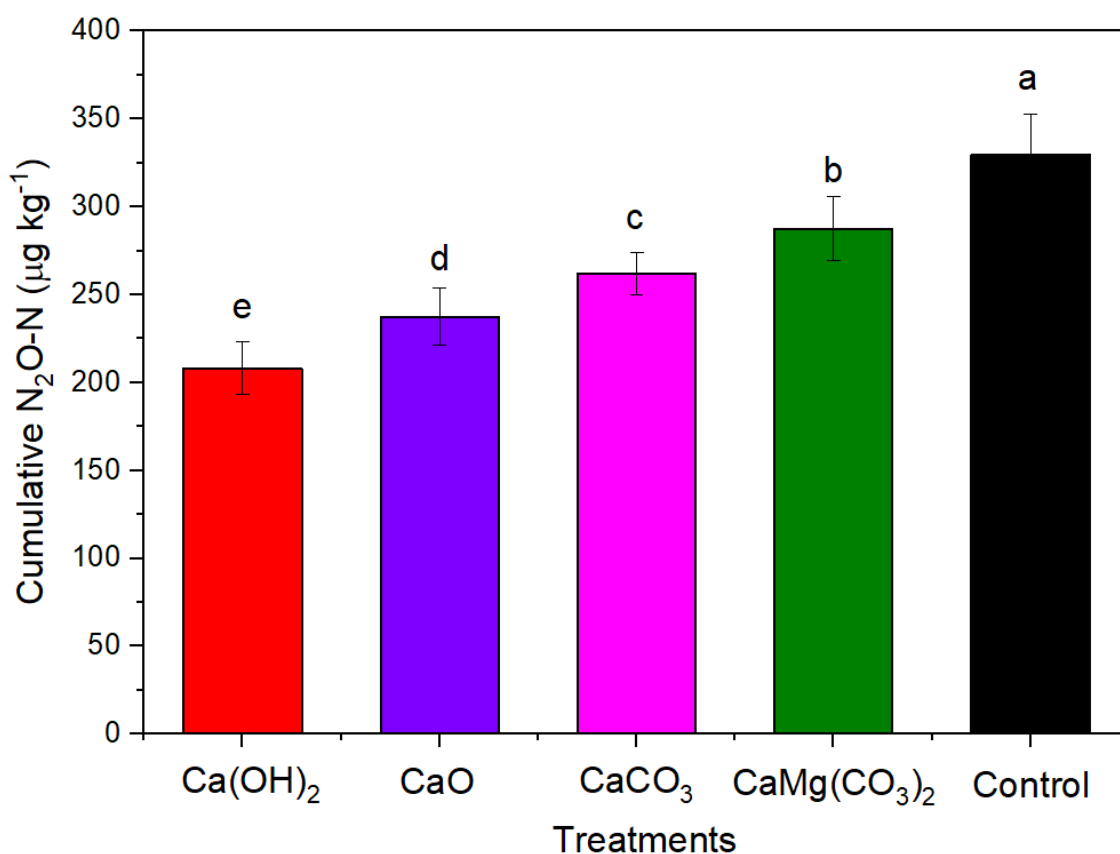


Figure 8. Cumulative soil N₂O emissions following the application of liming materials. Vertical bars denote the error bars of the means of three replicates. Different letters (from *a* to *e*) denote significant differences ($p \leq 0.05$) among the means of treatments.

4. Discussion

Acidic soils are generally considered as less efficient for high crop production. To ameliorate acidic soils, farmers usually amend them with liming materials. Application of liming materials has dual benefits of raising soil pH as well as supply essential elements, mainly Ca and Mg. In the present study, the liming materials used were Ca(OH)₂, CaO, CaCO₃, and CaMg(CO₃)₂. Application of all these liming materials obviously influenced N₂O emissions, but the magnitudes of the N₂O emissions dramatically altered with soil pH. In fact, high N₂O emissions were observed at low pH levels (without lime application) in the acidic soil in the present study.

High magnitudes of N₂O emissions from low pH soils can be explained by incomplete denitrification and less activity or complete inhibition of N₂O-reductase. Nitrous oxide reductase (N₂O-R) is the sole enzyme of the denitrification process, which reduces N₂O to N₂ at neutral, near neutral, or above 7 pH [18]. Therefore, higher magnitudes of N₂O emissions are expected from soils at low pH relative to higher pH values because of the incomplete denitrification process [19,20]. In the present study, N₂O emissions were perceptibly mitigated by the application of all selected liming materials. However, the highest decline in cumulative as well as emission rates of N₂O occurred in the Ca(OH)₂ treatment, and this was possible due to the highest pH value. Kunhikrishnan et al. [21] also indicated that the pH value could prominently control N₂O production and emissions, and Bakken et al. [22] commented that the possible mechanism involved in low magnitudes of N₂O emissions in limed soils at high pH values was pertinent to the activities of N₂O-reductase. It has been shown that the application of liming materials improved the activities of N₂O-reductase for N₂O reduction [23], and magnitudes of soil N₂O emissions are unswervingly controlled by pH [8,24]. These studies demonstrated that N₂O-reductase was functional at higher pH

relative to low pH, which led to a complete denitrification process and low N_2O emissions at high pH levels.

Results of raising soil pH regarding the effect on N_2O emissions have been proposed by several researchers. Stevens and Laughlin [25] reported that raising the soil pH from 6.5 to 8 eminently reduced N_2O emissions. Qu et al. [26] reported that acidic soils produced higher magnitudes of N_2O emissions, whereas neutral pH soils showed less magnitudes of N_2O emissions. Khan et al. [27] found that the application of calcium hydroxide to soil at the dose of 5.63 g kg^{-1} soil significantly decreased N_2O emissions by increasing soil pH from 5.2 to 7.6. Additionally, an 80-day laboratory study revealed that $\text{Ca}(\text{OH})_2$ amendment (1.1 to 5.6 g kg^{-1} soil) substantially reduced N_2O emission [28]. Moreover, some other experiments showed the following: application of $\text{Ca}(\text{OH})_2$ at the dose of 7.3 g kg^{-1} soil mitigated cumulative emissions of N_2O from 547 g ha^{-1} to 46 g ha^{-1} in a soil with a pH of 4.71 [29]. A 2-year research showed that increasing the pH from 4 to 5.5 by CaCO_3 application dwindled N_2O emissions from $0.96 \text{ mg m}^{-1} \text{ d}^{-1}$ to $0.88 \text{ mg m}^{-1} \text{ d}^{-1}$ [30]. The mitigation of N_2O emissions from limed soils showed that pH plays an imperative role in regulating such N_2O release to the atmosphere [31].

In the present study, the addition of liming materials greatly impacted mineral N concentrations displaying a quick decline of NH_4^+-N with time, indicating that the nitrification process sped up, as linked to the concurrent rise of NO_3^--N concentrations. Higher NO_3^--N concentrations at relatively higher soil pH levels advocate that microbes consumed N_2O as an electron acceptor instead of NO_3^--N . It can be observed from these results that complete denitrification occurred, rendering N_2O to N_2 conversion in all liming material treated soils, and thus correspondingly, low magnitudes of N_2O emissions occurred. Moreover, it is interesting to report herein that the trend and behavior of N_2O release from liming material amended soils corresponded with the changes in NH_4^+-N and NO_3^--N concentrations, but the degree of the mitigation of N_2O emissions did not follow the same pattern. The most rapid changes of mineral N dynamics were observed in the CaCO_3 treatment, whereas the highest reduction of N_2O emissions occurred in the $\text{Ca}(\text{OH})_2$ treatment. The discrepancies between the degrees of N_2O emission magnitudes following liming material application is plausible because of the potential of soil pH manipulation.

In addition to mineral N dynamics, the application of all liming materials influenced dissolved organic C, which is believed to be a readily available C substrate for microbial growth prolongation and proliferation, leading to processing nitrification and denitrification producing N_2O [32]. It is interesting to note that the changes in MBC comparing the end values with the starting values: ca. -10 mg/kg soil for the control versus ca. $+10 \text{ mg/kg}$ for treatment $\text{Ca}(\text{OH})_2$. Dissolved organic C acted as a substrate for microbes, conjecturing that available C favored N_2O reduction. Furthermore, high contents of MBC in the liming material added soils were detected when compared to the control, which indicated the likely high reduction of N_2O emissions.

5. Conclusions

The present research showed that the application of liming materials reduced magnitudes of N_2O emissions. The pronounced and maximum reduction of soil N_2O emissions occurred in the $\text{Ca}(\text{OH})_2$ treatment through increasing soil pH when compared to the other liming materials tested. The results suggest that N_2O emission mitigation is more dependent on soil pH than on C and N dynamics when capering different liming materials. Moreover, ameliorating the soil acidity condition is a promising option to alleviate N_2O emissions from acidic soils. The results can be considered of environmental relevance, and further research in this regard could be interesting, especially in the current context of global warming due to a variety of greenhouse gases released to the atmosphere from different compartments and due to various anthropogenic activities.

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